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Development of an Integrated *in-situ* Remediation Technology

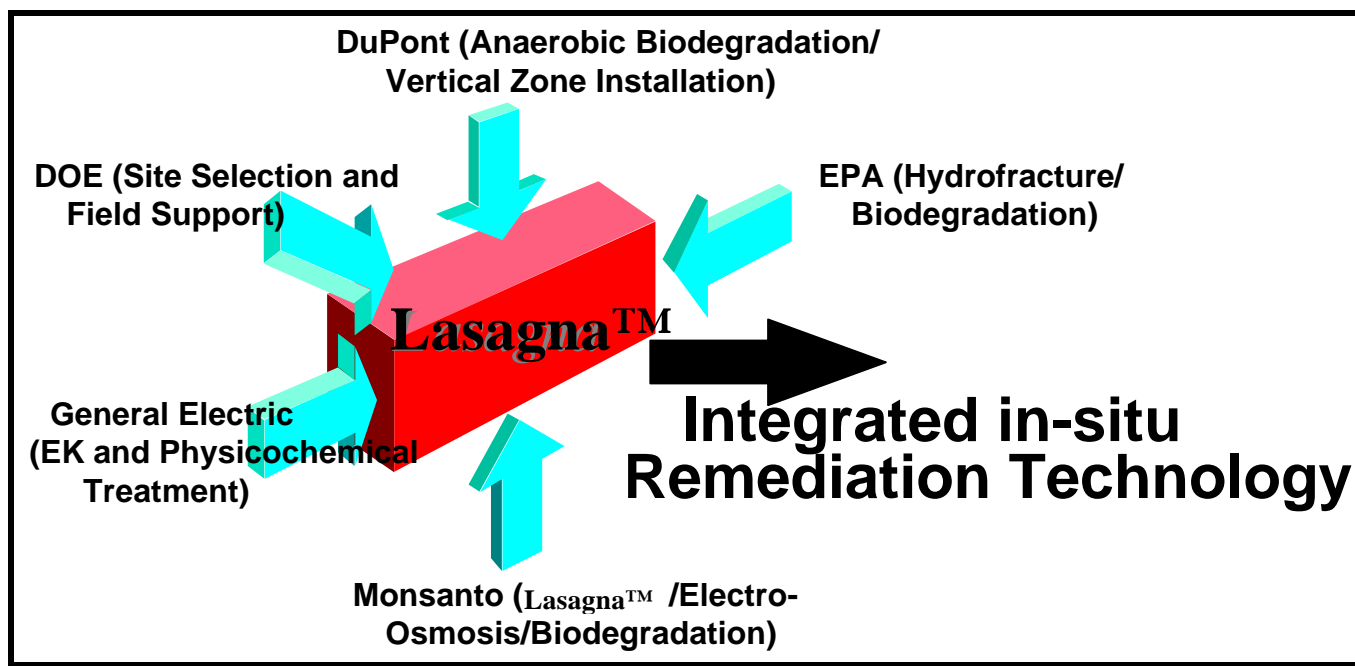
Topical Report for *Tasks #12 and 13 entitled "Large Scale Field Test of the Lasagna™ Process"* (September 26, 1994 - May 25, 1996)

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20 March, 1997

Re: Ordering Information for "*Development of an Integrated in-situ Remediation Technology*"
Topical Reports generated under DOE contract number DE-AR21-94MC31185 which was
signed September 26, 1994.

Dear Sir/Ms.:

The following table summarizes ordering information for all technical reports written for the above referenced contract. Copies of these reports may be obtained from the Office of Scientific and Technical Information [(423)576-8401] if you are with DOE or a DOE contractor, or from:

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(continued on next page)

Title	Document Number
<ul style="list-style-type: none"> • Topical Report for Task #1 entitled "Evaluation of Treatment Zone Formation Options" (September 26, 1994 - May 25, 1996) Stephen H. Shoemaker, Richard C. Landis, Ronald J. Griffith, Dale S. Schultz, and Gary E. Quinton (DuPont Company) 	DOE/METC/31185 —5436, DE97002165
<ul style="list-style-type: none"> • Topical Report for Tasks #2-4 entitled "Electrokinetic Modeling" (September 26, 1994 - May 25, 1996) Andrew P. Shapiro (General Electric Company) 	DOE/METC/31185 —5391, DE97002135
<ul style="list-style-type: none"> • Topical Report for Task #5 entitled "Cost Analysis" (September 26, 1994 - May 25, 1996) Gary Quinton, Dale Schultz, Richard Landis, Ronald Griffith, and Stephen Shoemaker (DuPont Company) 	DOE/METC/31185 —5389, DE97002134
<ul style="list-style-type: none"> • Topical Report for Task #6 entitled "Lab-Scale Development of Microbial Degradation Process" (September 26, 1994 - May 25, 1996) J. Martin Odom (DuPont Company) 	DOE/METC/31185 —5388, DE97002130
<ul style="list-style-type: none"> • Topical Report for Task #7 entitled "Development of Degradation Processes" (September 26, 1994 - May 25, 1996) M. J. Brackin, M. H. Heitkamp and S. V. Ho (Monsanto Company) 	DOE/METC/31185 —5495, DE97002165
<ul style="list-style-type: none"> • Topical Report for Tasks #8 and 10 entitled "Laboratory and Pilot Scale Experiments of the <i>Lasagna</i>TM Process" (September 26, 1994 - May 25, 1996) Sa V. Ho, Christopher J. Athmer, and P. Wayne Sheridan (Monsanto Company) and Andrew P. Shapiro (General Electric Company) 	DOE/METC/31185 —5375, DE97002150
<ul style="list-style-type: none"> • Topical Report for Task #9-Part I entitled "TCE Degradation Using Non-Biological Methods" (September 26, 1994 - May 25, 1996) Andrew P. Shapiro, Timothy M. Sivavec, and Sunita S. Baghel (General Electric Company) 	DOE/METC/31185 —5392, DE97002133
<ul style="list-style-type: none"> • Topical Report for Task #9 - Part II entitled "TCE Degradation Using Non-Biological Methods" (September 26, 1994 - May 25, 1996) Robert G. Orth and David E. McKenzie (Monsanto Company) 	DOE/METC/31185 —5393, DE97002131

(continued on next page)

<ul style="list-style-type: none">• Topical Report for Task #11 entitled "Evaluation of TCE Contamination Before and After the Field Experiment" (September 26, 1994 - May 25, 1996) B. Mason Hughes, Sa V. Ho, Christopher J. Athmer, and P. Wayne Sheridan (Monsanto Company) Stephen H. Shoemaker and John R. Larson (DuPont) Jay L. Clausen (LMES) and John L. Zutman (ORNL-Grand Junction)	DOE/METC/31185 —5496, DE97002166
<ul style="list-style-type: none">• Topical Report for Tasks #12 and 13 entitled "Large Scale Field Test of the <i>Lasagna</i>TM Process" (September 26, 1994 - May 25, 1996) Christopher J. Athmer, Sa V. Ho, B. Mason Hughes, P. Wayne Sheridan, and P. H. Brodsky (Monsanto Company) Andrew P. Shapiro, Roy F. Thornton, and Joseph J. Salvo (General Electric Company) and Dale S. Schultz, Richard C. Landis, Ron Griffith, and Stephen H. Shoemaker (DuPont)	DOE/METC/31185 —5390, DE97002156

A. Executive Summary

Development of an Integrated *in-situ* Remediation Technology

DOE Contract Number: DE-AR21-94MC31185

Topical Report for *Task #12 and 13 entitled:* ***“Large Scale Field Test of LasagnaTM Process”***

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Abstract: Contamination in low permeability soils poses a significant technical challenge to in-situ remediation efforts. Poor accessibility to the contaminants and difficulty in delivery of treatment reagents have rendered existing in-situ treatments such as bioremediation, vapor extraction, pump and treat rather ineffective when applied to low permeability soils present at many contaminated sites. This technology is an integrated in-situ treatment in which established geotechnical methods are used to install degradation zones directly in the contaminated soil and electroosmosis is utilized to move the contaminants back and forth through those zones until the treatment is completed. This topical report summarizes the results of the field experiment conducted at the Paducah Gaseous Diffusion Plant in Paducah, KY.

The test site covered 15 feet wide by 10 feet across and 15 feet deep with steel panels as electrodes and wickdrains containing granular activated carbon as treatment zones. The electrodes and treatment zones were installed utilizing innovative adaptation of existing emplacement technologies. The unit was operated for four months, flushing TCE by electroosmosis from the soil into the treatment zones where it was trapped by the activated carbon. The scale up from laboratory units to this field scale was very successful with respect to electrical parameters as well as electroosmotic flow. Soil samples taken throughout the site before and after the test showed over 98% TCE removal, with most samples showing greater than 99% removal. This high degree of removal is remarkable since some of the pre-test soil samples indicated the presence of residual pure phase TCE (DNAPL). Analysis of the carbon samples retrieved from various treatment zone locations showed that overall about 50 percent of the TCE can be accounted for, which is quite good considering the difficulty in handling TCE and the highly non-uniform distribution of TCE in the soil. Monitoring TCE levels in the air during the test showed that only about 4% of the total TCE could have been lost through evaporation. As far as we know, this is the first time that an organic contaminant could be cleaned up so uniformly from an actual contaminated clayey site. The Lasagna™ process shows great promise as an in-situ soil remediation technology, especially for low permeability soils.

Special Acknowledgments

Over the past two years, considerable technical and non-technical support has been provided by a large group of government, university, and industry scientists. Although it is impossible to name all of them at this time, special thanks should be extended to a number of organizations.

The Environmental Protection Agency's Remediation Technology Development Forum was instrumental in providing an environment where diverse groups could meet and discuss common issues that could result in drastic reductions in the costs of environmental remediation.

Scientists and consultants from Battelle Pacific Northwest Laboratories, Department of Energy (EM-531 and METC), Lockheed Martin Energy Systems, Environmental Protection Agency (RREL), University of Cincinnati, Clean Sites, Inc., and the Lasagna(TM) Consortium provided valuable input to the planning, funding, and execution of this field demonstration of the Lasagna(TM) process.

The Lasagna(TM) Consortium would like to thank all of these individuals for providing critical resources for the successful planning and execution of this study.

B. Acronyms and Abbreviations

A	Area (cross sectional)
C4-7	Carbon wick sample -4th row from anode, 7th wick.
C _{smax}	maximum adsorbed TCE concentration
DC	direct current
DNAPL	dense non-aqueous phase liquid
DOE	Department of Energy
e-	electrons
E _o	electrochemical reduction potential
EO	electro-osmosis
EPA	Environmental Protection Agency
ESC	Monsanto's Environmental Sciences Center
FIDAP	mass and heat transfer fluid model
GAC	granular activated carbon
GC-EC	gas chromatograph with electron capture detector
GE	General Electric
i _e	voltage gradient
K	Langmuir isotherm adsorption constant
k _e	coefficient of electroosmotic permeability
L17-08	soil core number
MMES	Martin Marietta Energy Systems
OH ⁻	hydroxide anion
ORNL	Oak Ridge National Laboratory
PC	personal computer
PGDP	Paducah Gaseous Diffusion Plant
pH	measure of acid content
Q	volumetric flow rate
ROA	Research Opportunity Announcement
TCE	trichloroethylene
WKWMA	Western Kentucky Wildlife Management Area

C. Units

°C	Celsius, degrees Celsius
cm	centimeters
g, gm	grams
hr	hours
kg	kilograms
L	liters
mg	milligrams
mA	milliamps
sec	seconds
μs	microseconds
V	volts
wt%	percent by weight

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E. Background

Statement of the Problem

Contamination in low permeability soils poses a significant technical challenge to *in-situ* remediation efforts. Poor accessibility to the contaminants and difficulty in delivery of treatment reagents have rendered existing *in-situ* treatments such as bioremediation, vapor extraction, and pump and treat, rather ineffective when applied to low permeability soils present at many contaminated sites.

The Solution

The proposed technology combines electro-osmosis with treatment zones that are installed directly in the contaminated soils to form an integrated *in-situ* remedial process. Electro-osmosis is an old civil engineering technique and is well known for its effectiveness in moving water uniformly through low-permeability soils with very low power consumption.

Conceptually, the integrated technology could treat organic and inorganic contamination, as well as mixed wastes. Once developed, the technology will have tremendous benefits over existing ones in many aspects including environmental impacts, cost effectiveness, waste generation, treatment flexibility, and breadth of applications.

Consortium Description

A Consortium has been formed consisting of Monsanto, E. I. du Pont de Nemours & Co., Inc. (DuPont) and General Electric (GE), with participation from the Environmental Protection Agency (EPA) Office of Research and Development and the Department of Energy (DOE) Environmental Management Office of Science and Technology. The five members of this group are leaders in their represented technologies and hold significant patents and intellectual property which, in concert, may form an integrated solution for soil treatment. The Consortium's activities are being facilitated by Clean Sites, Inc., under a Cooperative Agreement with EPA's Technology Innovation Office. A schematic diagram of the government/industry consortium is shown on the front page of this topical report.

Management Plan

A Management Plan for this project was prepared by Monsanto and submitted on November 30, 1994. That plan summarized the work plan which was developed in conjunction with DuPont, GE, EPA's Risk Reduction Engineering Laboratory (RREL), Martin Marietta Energy Systems (MMES), and the Department of Energy. The DOE Gaseous Diffusion Plant in Paducah, Kentucky, was chosen as the site for the initial field tests.

CDM Federal Programs Corporation was chosen to provide the on-site support of the field tests which were installed at the DOE site in November 1994. This experiment tested the combination of electro-osmosis and *in-situ* sorption in the treatment zones. In 1994 and 1995, technology development was carried out under the present contract by Monsanto, DuPont, and GE. These studies

evaluated various degradation processes and their integration into the overall treatment scheme at bench and pilot scales.

Technical Deliverables

Tables E1 and E2 summarize the 13 technical tasks and the 8 topical reports which will be written describing the results obtained in the technical tasks. These two tables show which organization is primarily responsible for the tasks and for preparing the topical reports. The present topical report summarizes the results of tasks 12 and 13.

Table E1. List of Tasks and Responsible Company

Task	Company
Task 1 - Evaluation of Treatment Zone Formation Options	DuPont
Task 2 - Electrokinetic Model Validation and Improvement	GE
Task 3 - Design Guidance for Field Experiments	GE/DuPont
Task 4 - Analysis of Electrode Geometry and Soil Heterogeneity	GE/DuPont
Task 5 - Cost Analysis	DuPont/Monsanto
Task 6 - Lab-Scale Development of Microbial Degradation Process	DuPont
Task 7 - Lab-Scale Electrokinetic and Microbial Degradation	Monsanto
Task 8 - Lab-Scale Tests of Lasagna Process Using DOE Paducah Soil	Monsanto
Task 9 - TCE Degradation Using Non-Biological Methods	GE/Monsanto
Task 10 - Bench- and Pilot-Scale Tests	Monsanto
Task 11 - Establish Contamination Conditions Before and After Tests	Monsanto/DuPont/ MMES
Task 12 - Design and Fabrication of Large-Scale Lasagna Process	Monsanto/DuPont/ Nilex
Task 13 - Large-Scale Field Test of Lasagna Process	Monsanto/CDM

Table E2. List of Topical Reports and Responsible Company

Topical Report	Company
Task 1 - Evaluation of Treatment Zone Formation Options	DuPont
Tasks 2 - 4 Electrokinetic Modeling	GE
Task 5 - Cost Analysis	DuPont
Task 6 - Laboratory-Scale Microbial Degradation	DuPont
Tasks 7, 8, 10 - Bench- and Pilot-Scale Tests of Lasagna Process	Monsanto
Tasks 9 - TCE Degradation Using Non-Biological Methods	GE
Task 11 - Contamination Analysis, Before and After Treatment	Monsanto
Tasks 12 and 13 - Large-Scale Field Test of Lasagna Process	Monsanto

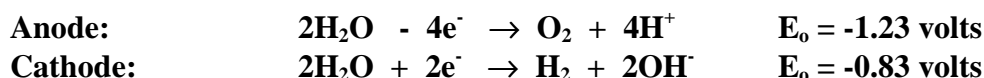
F. Topical Report of Tasks #12 and 13

INTRODUCTION

Contamination in low permeability soils poses a significant technical challenge to *in-situ* remediation efforts. Poor accessibility to the contaminants and difficulty in delivery of treatment reagents have rendered existing *in-situ* treatments such as bioremediation, vapor extraction, and pump and treat rather ineffective when applied to low permeability soils present at many contaminated sites.

A novel, *in-situ* technology aimed at cleaning up contamination in heterogeneous or low-permeability soils is being developed. The new approach involves the synergistic combination of electro-osmosis with other technologies. Used since the 1930s for dewatering clays, silts, and fine sands (1), electro-osmosis (EO), or more generally electrokinetics, has recently received increasing attention as an *in-situ* method for soil remediation (2-7). Water injected into the soil at the anode flows by electro-osmosis through the contaminated soil, bringing the contaminants (metals, organics) to the surface at the cathode region for further treatment or disposal.

Advantages with electro-osmosis include uniform water flow through heterogeneous soil, high degree of control of the flow direction, and very low power consumption. As currently practiced, however, the technology suffers several limitations associated with electro-osmosis for large-scale remedial applications. These include low speed (liquid flow induced by electro-osmosis typically moves about 1 inch per day for clay soils), additional aboveground treatment, unstable long-term operation resulting from soil drying and cracking, steep pH gradient in the soil bed, and precipitation of metals and minerals near the cathode (4,5,7). The pH changes primarily result from water electrolysis as the predominant electrode reactions, generating acid at the anode and base at the cathode according to the following reactions:



where E_0 is the standard electrochemical reduction potential. Recent efforts to mitigate the pH problem primarily involve conditioning the anode and cathode solutions through external recirculating loops (8,9).

Monsanto's new approach is an integrated *in-situ* treatment in which established geotechnical methods are used to install degradation zones directly in the contaminated soil and electro-osmosis is utilized to move the contaminants back and forth through those zones until the treatment is completed. Conceptually, the integrated technology could treat organic and inorganic contamination as well as mixed wastes. The process has been given the nickname "*Lasagna*" due to the many layers created (10,11). Diagrams of the vertical and horizontal *Lasagna*TM process configurations are shown in Figure 1.

A consortium of industry (Monsanto, DuPont and General Electric) was formed in collaboration with the Department of Energy (DOE) and Environmental Protection Agency (EPA) to combine expertise and resources for accelerating the development of this technology. The collaboration led to a field experiment of the LasagnaTM process at a DOE site in Paducah, KY, that has clayey soil contaminated with trichloroethylene (TCE). The Paducah Gaseous Diffusion Plant (PGDP) site was chosen from a list of Department of Energy sites based on two major criteria: low-permeability soil and a single chlorinated contaminant without heavy metals or radionuclides. DOE contributed by providing the contaminated site in Paducah for the test, soil sampling and analysis support (through Martin Marietta), and funds through a Research and Opportunity Announcement (ROA) grant. The entire project, including laboratory research and field work was about one million dollars. CDM-Federal, Inc., was hired by the consortium to construct and manage the field experiment. The field unit was constructed in November and December 1994, began operating in January 1995, and was completed in May 1995. This report documents this Phase I LasagnaTM field experiment.

OBJECTIVES OF THE FIELD EXPERIMENT

The purpose of this field test was to experiment with the coupling of electro-osmotic removal of TCE from the contaminated soil with *in-situ* adsorption by activated carbon in treatment zones in the vertical configuration. This was the initial phase of the LasagnaTM project, to be followed by a second phase incorporating *in-situ* degradation of TCE, either chemically or biologically.

The major technical objectives for conducting the field test included studying the scaled-up characteristics of key operating parameters as well as the effectiveness of TCE removal. Some important operating issues involved:

- * design, installation and cost of treatment zones
- * electrical effects of voltage, current, power, soil conductivity, heating, etc.
- * electrokinetic effects such as electro-osmotic flow, pH profile, solution conductivity, responses to polarity & flow reversal, and long-term operation

Issues related to TCE removal included:

- * extent of soil cleanup
- * effectiveness of carbon adsorption in an electro-osmotic environment
- * overall mass balance.

Field data were also needed for the development of a mathematical model of the process and for refining the estimate of remediation cost.

MATERIALS & METHODS

SOIL AND CARBON ANALYTICAL PROCEDURES

Soil sampling was carried out by Oak Ridge National Laboratory (ORNL) under the direction of Martin Marietta Energy Systems (MMES). TCE was extracted from the soil with hexane. Soil analysis was performed at the site using a gas chromatograph equipped with an electron capture detector (GC-EC). Carbon sampling was performed by ORNL and ESC. Carbon analysis was performed at the Monsanto Lab in St. Louis using GC-EC. The TCE was extracted from the carbon using hexane. Details of the sampling and analytical protocols and methods can be found in the Topical Report for Task#11 report issued by Monsanto.

EXPERIMENTAL SETUP

The location of the test site at the Paducah plant is depicted in Figure 2. The field test covered a soil section 15 feet long by 10 feet wide and approximately 15 feet deep.

Electrodes and Treatment Zones Setup: The electrode zones consisted of 8 electrode wicks side by side at each end of the field unit installation. There were a total of 4 treatment zones installed consisting of 11 or 12 individual wicks placed side by side in a staggered configuration. The treatment zones were installed with 21 inches of soil between each zone. Figure 3 shows the layout of the field unit wicks. Two wicks in each row contained a special sampling cassette 6 inches wide. A stainless steel shell with 50 percent open area was constructed to hold a 6 inch wide section of wick drain. These cassettes could be retrieved during or at the end of the experiment for TCE analysis.

A control wick was installed at the west end of the unit and isolated hydraulically by a wall of sheet piling surrounding 3/4 the perimeter of the control zone. The control zone wick was constructed with a sampling cassette.

Emplacement: The construction of the treatment zones and electrodes was overseen by DuPont and described in detail in Topical Report for Task #1. A novel installation technique was developed and carried out with the assistance of NILEX corporation of Englewood Co. The treatment zones were comprised of a series of individual "wicks" 18.5 inches wide and 15 feet long placed vertically next to each other to create a curtain 15 feet wide and 15 feet deep and 1 inch thick. The wick material was made from a roll of Hydraway® wick drain material manufactured by Monsanto. The wicks were cut to length, filled with granular activated carbon (FiltroSorb - 400® from Calgon) and encased in a geosynthetic filter fabric cover (Tygar® from Remay). Each wick also had a 15 foot length of 1/2" polyethylene tubing running the full length of the wick to allow water samples to be taken from any wick and for water level control in the electrode wicks. The electrode wicks also contained a section of 1/4 inch thick plate of carbon steel 15 feet long and 15 inches wide.

The wicks were installed using a modified sheet piling installation technique. A 10 ton vibratory hammer mounted on a 25 foot high mast tower was connected to a steel mandrel sleeve that

moved vertically. A steel shoe was designed to open the hole and allow the mandrel to penetrate through the gravel and clay layers to a depth of about 15 feet below grade. The wick was then inserted into the mandrel and the mandrel was removed leaving the shoe and wick in place. A total of 46 treatment wicks and 16 electrode wicks were installed.

Water Management System: The fluid circulation was handled as depicted in Figure 4. Individual overflow tubes are inserted each into a cathode panel down to about the clay level. A pump was used to lift liquid from the cathode compartments into the cathode collection tank, which overflows into the anode supply tank through the electromagnetic flowmeter. Another pump was used to circulate the liquid on the anode side through the anode tank. Liquid levels were controlled using two 8-head peristaltic pumps. Each pump was used to siphon the water in the electrode wicks; one 8-head pump for the anode, one for the cathode. Siphon tubes were placed in each electrode wick to a depth of 5 feet below grade to control the water level at that point. Each pump exhaust went to a collection tank. The cathode collection tank fed the anode collection tank by gravity through a flow meter. Since electro-osmotic flow moves from anode to cathode, the cathode tank filled and drained to the anode tank at whatever the electro-osmotic flow rate happens to be. The anode collection tank was equipped with a flushing system which allowed the water to drain back to the anode wicks.

To help keep the water handling systems from freezing and to help secure the area, a temporary plastic enclosure was constructed over the test site including the water tanks. The air discharge from the cooling radiator for the electrical generator was used to blow warm air into the enclosure. A vent was placed at the opposite corner to allow air to leave the enclosure. This vent was later used to monitor TCE volatilization.

Power Supply and Data Acquisition System: Power to the test site was supplied with a diesel-run Kubota Power Generator GV-3190Q rated at 18 kw. Two Sorenson 300 volt, 35 amp power supplies provided the DC power to the field unit. The power supplies were connected in parallel to provide the required current.

A total of 12 thermocouples and 7 multi-depth voltage probes were installed in the unit. The thermocouples were installed at various depths and locations to monitor the temperature rise in the soil due to resistive heating. Each voltage probe measured the voltage potential at 2 foot intervals vertically. Figure 5 shows the layout of the voltage and temperature probes. A PC based data acquisition system was installed to monitor the operation of the unit. Twelve channels were monitored by the data acquisition system: 8 temperature points (two T3 points in the middle soil section, one at one of the electrodes, and one at the control area), total voltage, current for each power supply, and total flow rate. Duplicate manual measurements were made periodically along with the voltage potentials and the pH and conductivity of the electrode fluids.

Electro-osmotic Permeability Calculation: The electro-osmotic permeability can be calculated using the following equation:

$$Q = k_e i_e A$$

where Q = volumetric flow rate by electro-osmosis (cm³/sec)
 k_e = coefficient of electro-osmotic permeability (cm²/volt-sec)
 i_e = voltage gradient applied across the soil mass (volt/cm)
 A = cross-sectional area perpendicular to the direction of flow (cm²)

Note that the electro-osmotic flow is proportional to the applied voltage gradient, and that the electro-osmotic permeability has the units of velocity over field strength (cm/sec over volt/cm). The cross sectional area was about 81 cm² for the standard laboratory unit, 2600 cm² for the pilot (bathtub), and 2.09×10^5 cm² for the field unit.

RESULTS & DISCUSSION

SITE & SOIL CHARACTERISTICS

The Paducah experiment site consisted of a 4 to 5 feet of a gravel and clay mixture over about 40 feet of TCE contaminated sandy clay loam. The hydraulic conductivity of the sandy clay loam is estimated to be about 1×10^{-7} cm/sec. The hydraulic conductivity of the gravel/clay overfill has not been determined.

Clean Soil: Prior to the field test, a load of clean soil was obtained from the Western Kentucky Wildlife Management Area (WKWMA) site near the Paducah Gaseous Diffusion Plant. This soil was judged similar to the contaminated soil at the Paducah plant. Table 1 summarizes some characteristics of this clean soil. It was characterized as a clay loam, and contained approximately 0.2% organic carbon. Due to its very low organic content, the soil adsorbs very little TCE. Figure 6 shows the TCE adsorption isotherm for the soil. The Langmuir fit of this data gives a C_{smax} (maximum adsorbed amount) of 4.5mg/kg and an adsorption constant, K , of 4.73 mg/L. Since the average TCE level in the contaminated soil at the test site was over 80 mg/kg, which translated to an average concentration in the pore water of almost 400 mg/L, most of the TCE at the site was probably not bound to the soil, and should readily move with the passing water. This clean soil was also used in the pilot (bathtub) unit for assessing long-term electro-osmotic performance and the heating effects.

Contaminated Soil: Four intact soil cores from the contaminated site were taken and analyzed by GE for moisture contents, wet and dry soil density. A summary of the results is shown in Table 2. The average wet soil density is 1.983 g/cm³, moisture content is 18.5 wt% of wet soil, and solid density is about 2.6 g/cm³. The electrical conductivity of the L17-08 core was measured as a function of temperature using the four-point method (12); it was found to increase steadily with temperature from 4°C to 70°C, shown in Figure 7 along with the fitted equation.

Electro-osmotic characteristics of the Paducah soil were studied in the laboratory with the sections of contaminated soil core samples re-packed in a standard, glass electro-osmosis unit (9 cm inside diameter, 14.5 cm in length for the soil column). Steel plates were used as electrodes

and four activated carbon zones, each 1 cm thick and 3 to 3.5 cm apart, were packed with the soil. For an applied voltage of 10 V, the current stabilized overnight to 11 mA. The electro-osmotic permeability obtained was calculated to be $1.2 \times 10^{-5} \text{ cm}^2/\text{V-s}$, which is a typical value for clayey soils.

FIELD RESULTS

The field unit was constructed during November and December 1995. The experiment was started (power on) on January 3, 1995 and lasted for four months. The voltage was then reversed for about one week, primarily to collect voltage and current data, before the power was turned off. Key operating characteristics of the test are reported below.

Voltage and Current: The initial voltage was set at 138 volts and the corresponding current was approximately 41 amps. This also happened to be the maximum current a single generator could handle. With the current held constant at 40 amps, the voltage slowly decreased with time and stabilized at 105 volts after one month of operation. The lower voltage was due to the soil heating up (discussed below), which increased the electrical conductivity of the soil. The voltage gradient ranged from 0.45 to 0.35 volts/cm. The voltage drop across the unit was fairly linear and did not change over the course of the experiment (Figure 8). Total voltage and current profiles are shown in Figure 9.

The current distribution among the individual wicks at the anode and cathode were measured twice during the test. The first set of data was taken January 24 (3 weeks into the test) and shows an expected current distribution. The current through the end wicks was slightly higher than the center wicks. This was probably due to current leakage from the system around the edges of the unit. The second data set taken at the end of the test shows a slightly more random current distribution. The current difference is probably due to different corrosion rates of the individual wick electrode plates. Figure 10 shows the current distribution data.

Electrokinetic Effects: During the first month, an electro-osmotic flow rate of about 2 L/hr was measured, which was less than one half of the predicted value based on the bench-scale's measurements. This resulted from plugging of the cathode siphon tubes, causing water to "overflow" the cathode wicks. Once this problem was corrected, the flow rate averaged approximately 4 to 5 L/hr, corresponding to an electro-osmotic permeability of $1.2 \times 10^{-5} \text{ cm}^2/\text{V-s}$. This value, as well as the water flow rates, thus agreed very well with the lab scale data.

With the first month's flow rate corrected to a value of 4 L/hr, the unit moved three pore volumes of water (between adjacent treatment zones) over the 4 month operating period. Figure 11 shows the flow rate and the corresponding pore volumes during the field test. A spreadsheet of the recorded field data is included in Appendix A. Appendix B contains the logged data from the data acquisition system.

Conductivity and pH: The pH of the anode fluid decreased from near neutral at the beginning to between 5 and 6 for most of the test. This mild pH behavior for an electro-osmosis experiment was a direct consequence of iron corrosion rather than water electrolysis as the predominant anodic reaction, which had also been observed in laboratory units. During the last month of the test, however, the anode fluid pH dipped rather unexplainably down to 2-3. The pH of the cathode fluid increased rapidly to around 12 and stayed there for the whole experiment, which is normal for electro-osmosis due to the electrolysis of water generating OH^- at the cathode.

The conductivity of the anode solution ranged between 1000 to 3000 $\mu\text{S}/\text{cm}$ for most of the test, except near the end when it fluctuated widely with occasional spikes up to 25,000 $\mu\text{S}/\text{cm}$. The prime ion species were iron and chloride. This behavior was probably related to the low pH noted above during this period, which leads to solubilization of many salts. The cathode solution conductivity was fairly constant at around 10,000 $\mu\text{S}/\text{cm}$ with the primary ion species being sodium. These values were consistent with the laboratory data. A plot of pH and conductivity values over time are shown in Figure 12. At the end of the test, water samples were taken from every wick to determine the distribution of anions, cations, pH and specific conductance. A table of final water quality data is included in Appendix C.

Temperature Effects: The initial temperature of the soil at the 10-foot depth was 15°C. The temperature rise at various locations due to the electrical power is shown in Figure 13. The core temperature (hottest spot) reached a maximum of 45.2°C at the end of the test. Notice the control zone temperature also increased. A possible reason for the increase in temperature in the control zone is that stray current could flow through it, causing heating. Or it could be from thermal conduction due to the proximity of the control zone to the anode (less than one foot).

To predict temperature rises in field experiment, a mathematical model was developed by Andrew Shapiro of General Electric using FIDAP[®], a commercial computational fluid dynamics program. The governing equations included energy transport by conduction, electro-osmotic convection, and heat generation from Joule heating. Temperature dependencies of electrical conductivity and electro-osmotic permeability were accounted for. Specific examples simulating possible scenarios for the Lasagna[™] pilot test in Paducah, KY, were studied. Based on the lab k_e value and estimated thermal conductivity and heat capacity of the soil, the operating current was to be set to 40 amps, which would keep the temperature of the unit below 50°C. With the appropriate values for thermal conductivity and heat capacity of the soil, the model predictions correspond very well with the field data as shown in Figure 14.

Polarity Reversal: The polarity was reversed for 1 week at the end of the test to determine how the field unit would respond. The unit was operated at a reverse voltage of 90 volts and a current of 39 amps. The unit responded very similarly to the laboratory and "bath tub" size experiments. There was an increase in electrical conductivity of the soil when the polarity was reversed and the pH at the electrodes was shifting from high to low at the new anode and from low to high at the new cathode. These results indicate that polarity reversal can be readily utilized in the field, if needed for reversing flow or neutralizing pH and/or osmotic gradients.

Overall, the field unit behaved very much like the laboratory and pilot size experiments. The unit scale-up was almost exact. The electro-osmotic conductivity, pH and conductivity trends, power requirements, temperature trends and operational stability all were in agreement with laboratory results.

TCE REMOVAL AND MASS BALANCE

Pre-Test Soil Samples: Prior to the installation of the Lasagna™ field unit, soil samples were taken and analyzed by Oak Ridge National Laboratories (ORNL) personnel. A total of 12 bore holes were made with samples taken every 1 foot to a depth of 15 feet below ground surface. Nine bore holes were completed within the boundary of the Lasagna™ unit, two in the control area and one to the outside west of the unit. Figure 15 shows the locations of the bore holes with respect to the wicks. The results show TCE concentrations in the soil ranged from 1 mg/kg to over 500 mg/kg with an overall average of 83.3 mg/kg for the 12 bore holes from 4 to 15 foot depths (Table 3). The average concentration of TCE within the test site (15' x 15' x 10') is almost the same, 83.2 mg/kg, resulting in a total amount of TCE of about 9.25 kg.

Intermediate Carbon Samples: In order to assess the progress of the test, the carbon cassette from wick C-7 was removed for analysis after 2 pore volumes of water exchange had occurred. Carbon samples were taken every foot from 3 to 15 feet. TCE levels on the carbon were found to be quite high, ranging from several thousand to over 10,000 mg TCE/kg carbon (Figure 16). This indicates that TCE was being effectively flushed from the soil and trapped on the carbon. Also shown in Figure 16 are the additional amounts of TCE trapped on the fresh carbon inserted into cassette of wick C4-7 for the remainder of the test (1 additional pore volume). The data show that, except at the lowest depths, little additional TCE was trapped by the carbon, which indicates that the soil was probably clean of TCE.

Post-Test Soil and Carbon Samples: At the completion of the test (3 pore volumes total), all carbon sampling cassettes were removed and analyzed in the same manner as the intermediate samples. Soil samples were also taken. Twelve bore holes were dug near the original pre-test bore holes to a depth of 15 feet and slightly lower. Results in Table 4 show very high as well as uniform removal of TCE from the treated soil. The final soil concentrations were generally below 1 mg/kg, with an average of 1.1 mg/kg for the 9 bore holes within the test boundary. TCE removal ranged from 92.4 to 99.8%, with an overall average of 98.4%. The soil samples taken either outside or deeper than the test zone (below 15 feet) still showed substantial amounts of TCE present. TCE removals outside the test boundary were 26% and 51% from the two spots in the control zone, and 68% from the west of the Lasagna™ unit. The TCE removal in the control zone was probably due to diffusion into the carbon wick (see modeling below). The removal in the area to the west was probably due to both migration and diffusion from the anode area and/or sampling inaccuracies. Despite significant degrees of removal from the control areas, the definite contrast between TCE levels in the soils within the treatment area and without (outside or deeper)

is quite remarkable and shows conclusively the effectiveness of the process for cleanup of the contaminated soil.

Several of the pre-test soil samples showed TCE concentrations greater than 225 mg/kg (12 through 15 foot samples of borehole L-08). These values would indicate a residual DNAPL situation since a soil TCE concentration of 225 mg/kg corresponds to an equilibrium pore water concentration of 1100 mg/l - based on a bulk soil density of 2 g/cc, a porosity of 0.4 (see Table 1) and subtracting 4.5 mg TCE/kg soil from the total due to adsorption. In these likely DNAPLs locations, TCE levels were reduced to less than 1 mg/kg, except for the 15 foot deep sample which was reduced to 17.4 mg/kg. These results show that the process could be effective for removing residual DNAPL TCE from the soil.

TCE Mass Balance: Attempts were made to determine the mass balance for TCE taking into account the amounts in the soil before and after the test and the amount trapped on the carbon. This is a complex task due to the following reasons:

- a) TCE levels in soil varied in all directions, so an averaging process was needed.
- b) Not all the soil samples were taken directly in front of the corresponding carbon cassettes, so another estimating procedure was required.
- c) The carbon cassettes were installed at the site about a month before the experiment actually started. So some TCE from the soil was lost due to passive diffusion to the carbon cassette in the back row opposite the direction of electro-osmotic flow. Based on diffusion modeling (Table 7 below), the loss could be about 5%.
- d) Volatilization of TCE from the soil into the atmosphere did occur to some extent. Estimates calculated from measurements of the air samples (shown below) indicated that the volatilization loss was less than 5% of the total.
- e) Possible TCE degradation reactions.

Because of the above complications, the mass balance results ought not to be viewed as more than a gross indication of the fate of TCE.

Contour maps of TCE levels in the treated area were obtained by DuPont using a kriging method. The maps are shown in Appendix D. Table 5 shows the estimated TCE levels in soil in front of each carbon wick that contained a carbon cassette. Table 6 shows the mass balances obtained for various locations, each from 4 to 15 foot depth. As can be seen, the mass balance ranges from 20 to 80%, with an overall average of about 50%. These numbers are judged to be excellent considering the uncertainties mentioned above. A copy of the individual sample data is included in Appendix E.

Volatilization Test: To identify possible loss of TCE through volatilization, the air in the unit enclosure was sampled two times during the test using dosimeters passively exposed to the flowing air stream for about 48 hours. The forced air draft caused by the generator cooling system was measured to be 300 cubic feet per minute (CFM). The first sampling performed on

Jan 24, 1995 resulted in an emission rate of 2.3 gm TCE per day. The core temperature of the LasagnaTM unit was 29.5°C at this time. The second sampling was performed on March 2 when the core temperature of the unit was 35°C. The TCE emission rate was for that sampling was 3.2 gm per day. Dosimeters placed in the inlet air duct (from the generator) showed non-detectable amounts of TCE. An emission rate of 3.2 grams per day for a period of 4 months results in an estimated 4% loss of TCE through volatilization. Figure 17 shows the monitoring locations and the sampling results for the second test.

Role of Diffusion: Due to the significant removal of TCE in the control area, the effect of diffusion on the removal of TCE from the contaminated soil was modeled. The model assumes

- slab configuration of 22 in-thick soil mass bracketed with infinite sinks (zero TCE concentration in the carbon wick drain), which is essentially correct since the activated carbon used has very high capacity for TCE (500 mg/g carbon) compared to the amount of TCE in soil (equivalent to less than 15 mg TCE/g carbon).
- no retardation from soil interaction, a good assumption due to the very low adsorption of TCE (maximum amount adsorbed = 4.5 mg TCE/kg soil)

The problem is similar to the unsteady state heat conduction in slab. The differential equation for unsteady state diffusion of TCE from the soil matrix is

$$\frac{dC}{dt} = D_e \frac{d^2 C}{dx^2} \quad (1)$$

where $C(x,t)$ is the concentration of TCE in pore water at time t and position x
 t is the elapsed time

D_e is the effective diffusion coefficient for TCE in the pore water of the soil matrix.

With τ as the soil tortuosity, $D_e = D_{\text{bulk}}/\tau$

For diffusion in/out from both sides of the slab held at a constant surface concentration (C_s) and uniform initial concentration of TCE in the soil, the analytical solution (13) to equation 1 is

$$\frac{C_s - C_a}{C_s - C_o} = \frac{8}{p^2} \left(e^{-a_1 N_{Fo}} + \frac{1}{9} e^{-9 a_1 N_{Fo}} + \frac{1}{25} e^{-25 a_1 N_{Fo}} + \dots \right)$$

Where

- C_s : constant concentration at either surface of the slab
- C_a : average concentration in the soil at time t
- C_o : initial concentration of TCE in the soil matrix
- N_{fo} : Fourier number, defined as $D_e t/s^2$

$$s : \quad 1/2 \text{ of the slab thickness}$$

$$a_1 = (\pi/2)^2$$

The results are shown in Table 7a-b in percents of TCE removal from the soil for various spacing of two adjacent carbon treatment zones and for temperatures from 15 to 60°C. The effects of temperature on water viscosity and TCE diffusion coefficient are estimated as follows.

Cragoe's equation for water viscosity from 15 to 60°C:

$$\eta_{H_2O}(T^{\circ}C) = 1.0019 \times 10^{-4} \left[1.2348(20 - T^{\circ}C) - \frac{0.001467(T^{\circ}C - 20)^2}{(T^{\circ}C + 96)} \right]$$

Hayduk-Laudie equation (14) for TCE diffusivity:

$$D_b = 13.26 \times 10^{-5} \mu_{H_2O}^{-1.4} V_{TCE}^{-0.589}$$

where V_{TCE} is the molal volume of TCE at its normal boiling point and is equal to 101.5 cm³/gmol.

As can be seen in Table 7a for the 22 in carbon zones spacing, TCE removal by diffusion after four months is 20.3% at 15°C, 24.1% at 25°C, and 29.9% at 40°C. These levels are way below the removal levels obtained in the treated soil of the field test. It is interesting that after 10 years passive diffusion can remove over 90% of TCE in the soil at 15°C. However, larger and more realistic treatment zone spacings of 5 to 7 feet drastically reduce the amount of TCE removal by passive diffusion. As shown in Table 7b, after 10 years the TCE removal by diffusion alone is less than 50% for a 5-foot spacing, and about 35% for the 7-foot spacing.

CONCLUSIONS

The LasagnaTM process has been shown effective for removing TCE uniformly from the contaminated clayey soils at the DOE Paducah site. After 3 pore volumes exchange of water, TCE removal was over 98%, with most samples showing over 99% removal. The process also seems effective for removal of residual DNAPLs, thought to be present at the lower depths due to the high TCE concentrations found in the soil there.

The LasagnaTM process appears to scale up very well. Overall, the field unit behaved very much like the laboratory experiments. The electro-osmotic conductivity, pH and conductivity trends, power requirements, temperature trends and operational stability all were in good agreement with laboratory results.

The data obtained in this first LasagnaTM field experiment were also valuable in refining the mathematical model of the process and the economic estimates of the treatment cost.

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Table 1. Paducah WKWMA Clean Soil Characteristics

Soil Type:	Clay Loam
Sand Content:	22 Percent
Silt Content:	46 Percent
Clay Content:	32 Percent
Moisture Content:	15-18 Percent
Organic Carbon Content:	0.2 Percent
Cation Exchange Capacity:	13.4 meq/100 g
Porosity:	0.4
Bulk Density:	2.0 g/cc

Table 2. Paducah Contaminated Soil Cores Physical Data

Core#	pH (avg)	Conductivity (uS/cm)	Moisture (%)
L17-04	6.05	115	19.5
L17-06	6.80	105	20.0
L17-08	5.80	70	6.7
L17-10	7.20	315	22.4

Electroosmotic Conductivity	$1.2 \times 10^{-5} \text{ cm}^2/\text{v-sec}$
Hydraulic Conductivity (estimated)	$1 \times 10^{-7} \text{ cm/sec}$